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DETERMINATION OF FIBER ORIENTATION IN
SHORT FIBER COMPOSITES

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Abstract

Laser scattering techniques are described which permit the rapid determination of quantitative descriptors of the fiber orientation in short fiber composites. The technique is based on the use of reduced images of photomicrograph negatives in which the fiber image is small enough to act as a diffraction slit for a laser light source. Characteristic scattering patterns are obtained for fibers in various states of orientation. Calibration curves are obtained by the use of computer generated scattering masks of known orientation.

Determination of Fiber Orientation in Short Fiber Composites

Introduction

Recent developments in predicting the mechanical properties of short fiber composites have resulted in an increased recognition of the role of fiber orientation. The results of Jarzebski, et. al.[1] for sheet molding compounds and McGee and McCullough[2] for injection molding compounds have shown that the fiber orientation is one of the major factors in determining the mechanical properties of these composites. The more recent work by Mukhopadhyay and McCullough[3] has shown the same for the thermal expansion coefficients.

Until now the only method of determining fiber orientation has been a time consuming and costly process of counting fibers at various angles. For example, after sectioning and polishing the composite, photomicrographs were taken. Next, a collage of the entire composite had to be assembled, and the individual fibers manually counted and their orientation recorded. From this data it was possible to construct a histogram containing the required orientation information. This could then be used in the modeling equations to estimate the properties of the composite. Obviously this process is too tedious for routine use.

This report covers a new method of determining fiber orientation which eliminates the need to count the individual fibers. The use of a light scattering technique is described which produces scattering patterns which are characteristic of the fiber orientation. These patterns are analyzed to provide the necessary information on the fiber distribution. First, however, a brief review of the modeling equations is given to provide the background on the type of fiber orientation information required.

Modeling Review

A very useful set of computer algorithms has been developed by Jarzebski, et. al[1] and by McGee and McCullough[2] for estimating the mechanical properties of short fiber reinforced composite materials. One of the most useful aspects of this work is the incorporation of a parameter to describe the internal microstructure of the composite. Based on the earlier work of Wu and McCullough[4], a single parameter is used to characterize the fiber distribution. This parameter is related to the second moment of the distribution, but due to the different assumptions regarding the symmetry of the material, two different expressions are required for sheet molded materials and injection molded materials.

Jarzebski et. al.[1], in studying sheet molding compounds, assumed that the fibers must all lie in the plane of the material. They also assumed that the fiber distribution was symmetric around the primary axis. Based on these assumptions, they defined the orientation parameter as

$$f_p = 2 \langle \cos^2 \phi \rangle - 1$$

where

$$\langle \cos^2 \phi \rangle = \int_0^{\pi/2} n(\phi) * \cos^2 \phi \; d\phi$$

and $n(\phi)$ is the function describing the orientation of the fibers. It is important to note that it is not necessary to know the actual distribution function, $n(\phi)$, but only the second moment. Thus any method of determining f_p will suffice in determining the fiber distribution.

The value of f_p ranges from zero to one. An f_p value of zero corresponds to a material which is random in the plane. This state is often referred to as quasi-isotropic. An f_p value of one corresponds to a material in which all the fibers are aligned along the unique axis. A material with this orientation is transversely isotropic.

For injection molded materials, McGee and McCullough[2] assumed that the fibers were distributed on the surfaces of cones whose axes lay along the unique axis of the composite. This distribution results in a transversely isotropic

material. Under this three dimensional distribution the orientation parameter is given by

$$f_a = .5*[3<\cos^2\theta> - 1]$$

where

$$<\cos^2\theta> = \int_0^{\pi/2} n(\theta)*\sin\theta*\cos^2\theta \; d\theta$$

Once again it is not necessary to know the complete distribution function; only the second moment is required.

The range of f_a is from -.5 to one. An f_a value of -.5 corresponds to a material in which the fibers are randomly oriented in a plane perpendicular to the unique axis. An f_a value of zero indicates a composite which is truly isotropic in all directions. The case of aligned fibers is again represented by an f_a value of one.

In addition to the parameters f_p and f_a, the parameters g_p and g_a are required for the evaluation of the thermoelastic properties. These parameters are defined as

$$g_p = .20*[8*<\cos^4\phi> - 3]$$

and

$$g_a = 1.25*[<\cos^4\theta> - 1]$$

However, McCullough[5] has shown that to a good approximation

$$g_p = 2*f_p(7 - 2f_p)/[5(4 - 2*f_p)]$$

and

$$g_a = 3*f_a/(5 - 2*f_a)$$

Consequently, a knowledge of f_p and f_a is sufficient to characterize the role of fiber orientation in determining thermoelastic properties. In the following sections, a method using a light scattering technique is described which allows one to measure these parameters.

Scattering Technique

The use of light scattering to estimate fiber orientation in a composite material is dependent on the ability to produce a suitable diffraction grating since the composite is generally optically opaque. To obtain the required diffraction grating the composite was first sectioned and polished. The sample was then photomicrographed at a magnification of 50X. A single photograph of the region of interest was sufficient. The negative of the photomicrograph was then photographed and the resulting slide was used as the diffraction grating. The process used to obtain this type of diffraction grating is described in detail in Appendix I.

Once the scattering mask had been produced, a standard light scattering apparatus was used to obtain the diffraction pattern. This apparatus is shown schematically

in Figure 1. To test this process to insure that it would indeed produce scattering patterns which would characterize the degree of fiber orientation, diffraction masks from two injection molded samples were produced. A highly oriented sample and a less oriented sample were chosen, since the minimum requirement was to be able to distinguish these two cases. The photomicrographs of these two samples are shown in Figure 2.

The scattering patterns from the samples of Figure 2 are reproduced in Figure 3. From these patterns the difference between the two samples is apparent. The highly oriented sample produced a scattering pattern with two very narrow lobes while the scattering pattern from the less oriented sample had much broader lobes. This preliminary experiment confirmed that the technique was feasible and that further investigation was warranted.

The method of observing the scattering patterns is discussed in Appendix II while the techniques used to reproduce the photographs of the original samples and the scattering patterns are described in Appendix III.

Experimental

The final objective of this initial investigation was to determine if the light scattering method was sensitive to slight variations in the fiber orientation. Since it was impractical to obtain photomicrographs of samples of known

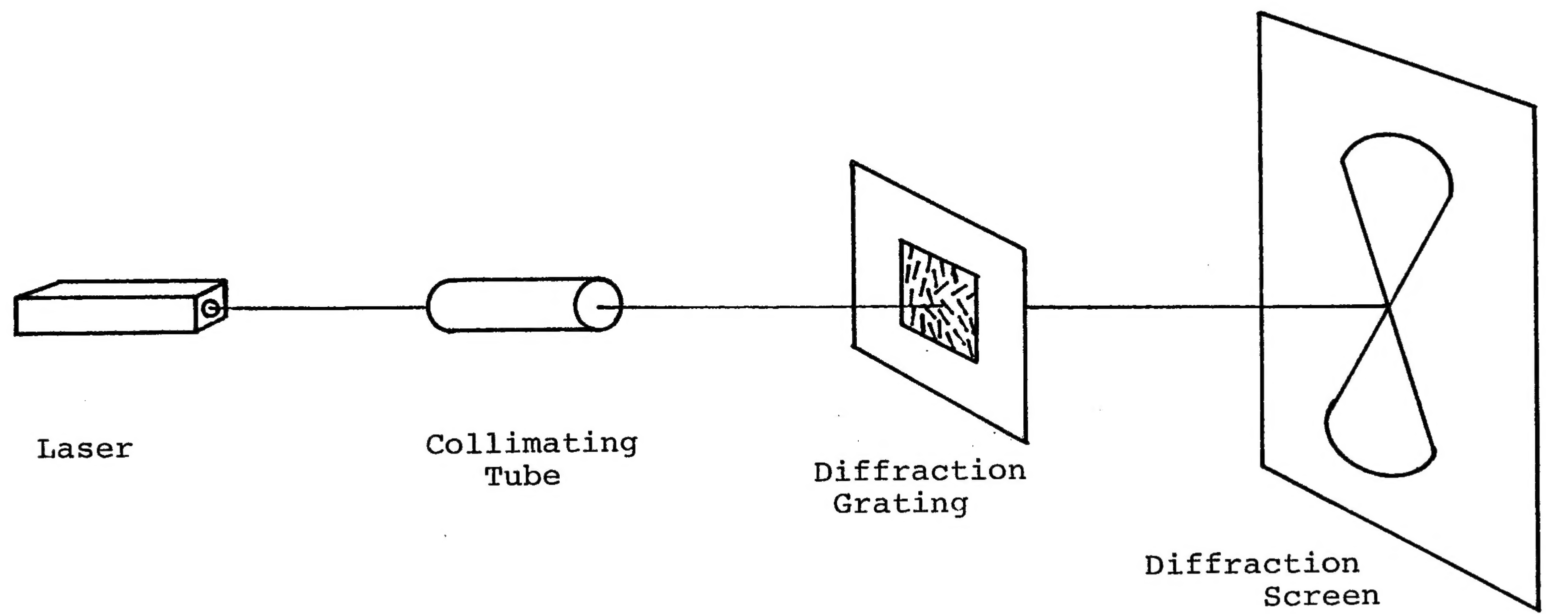
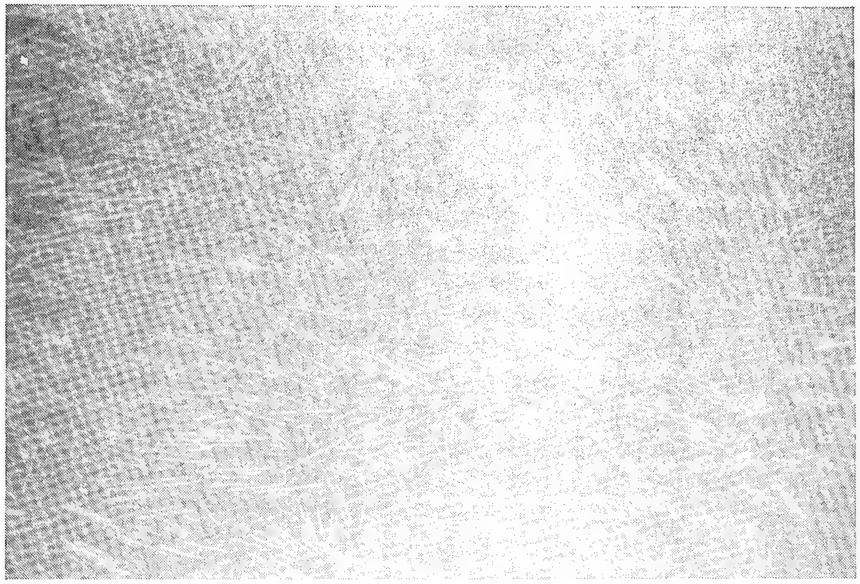
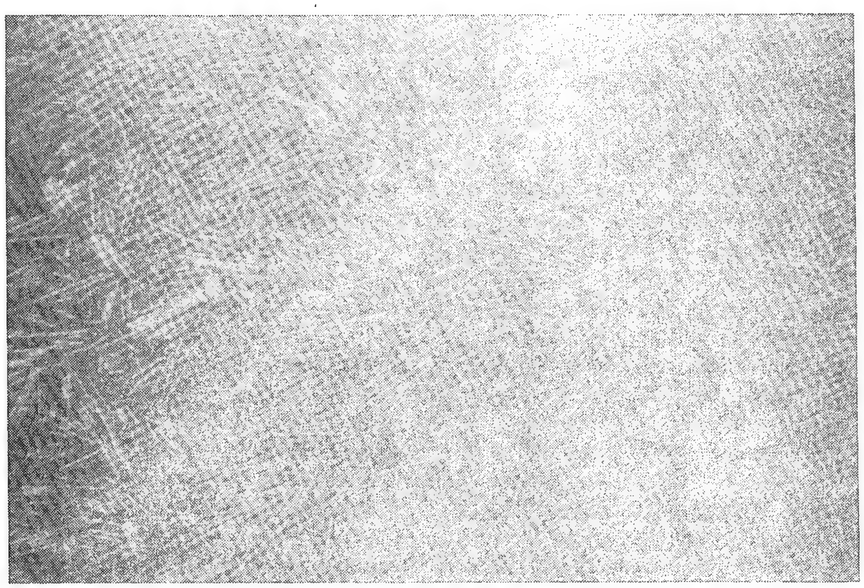


Figure 1. Schematic Diagram of Diffraction Apparatus

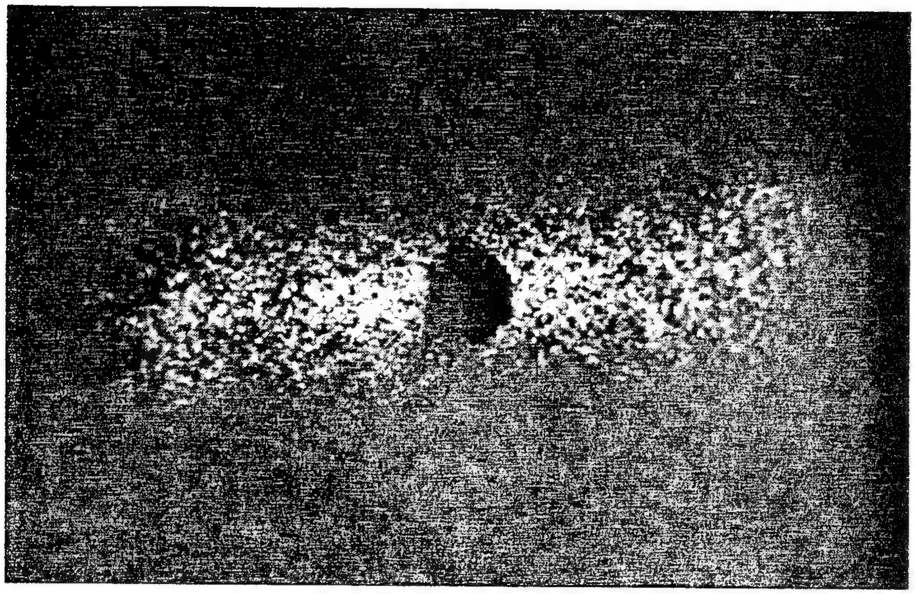


a) oriented

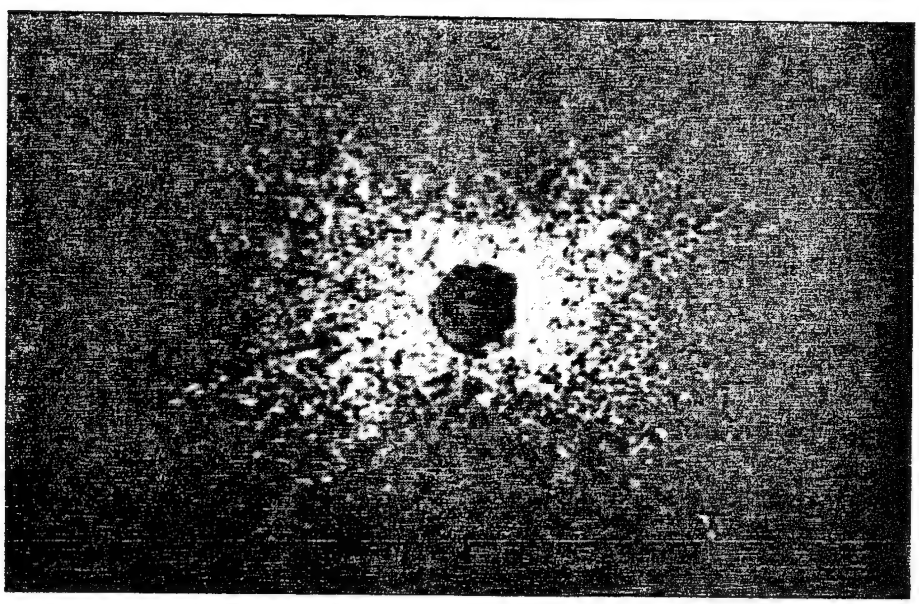


b) less oriented

Figure 2. Photomicrographs of Injection Molded Composites.

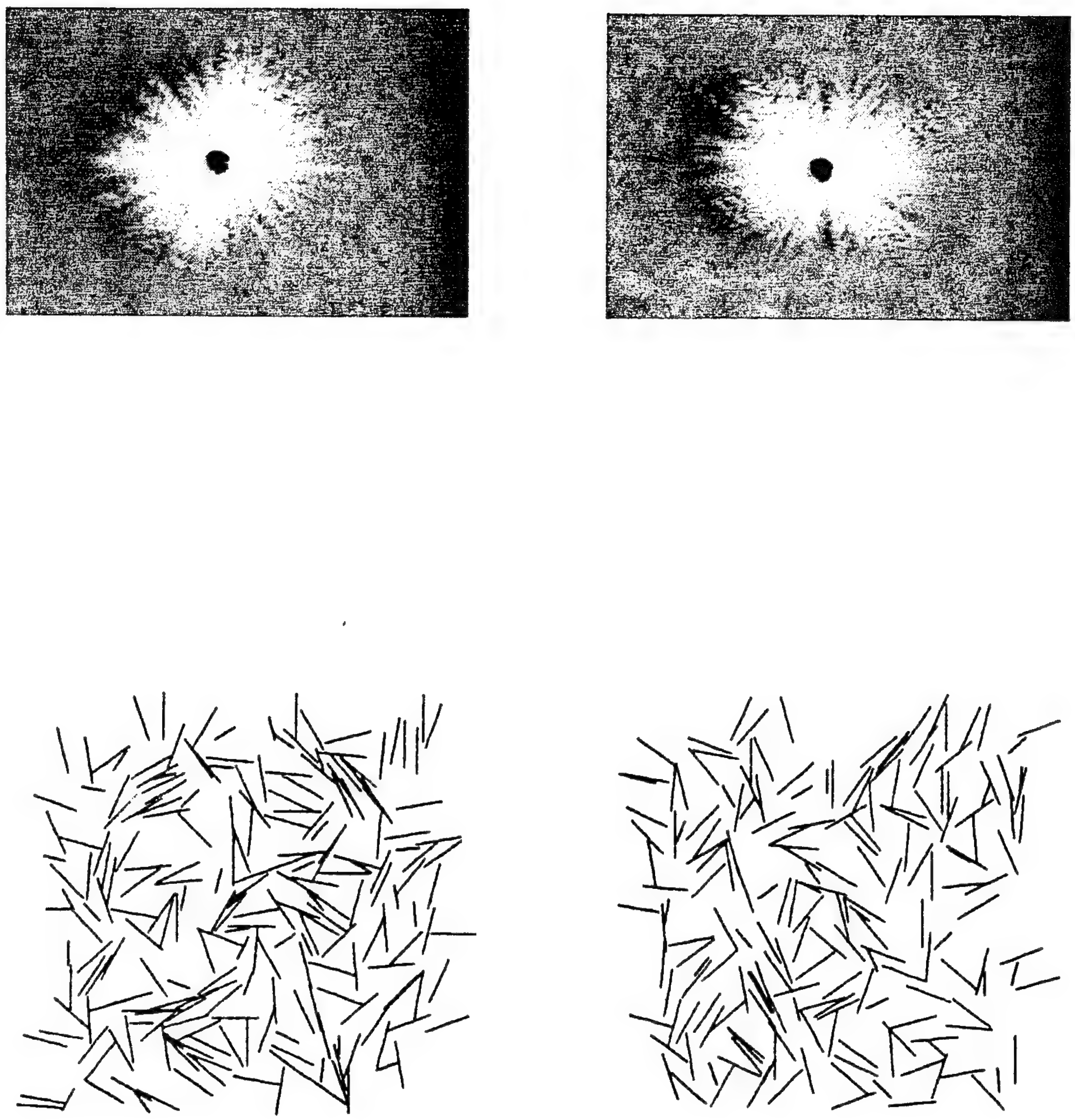


a) oriented



b) less oriented

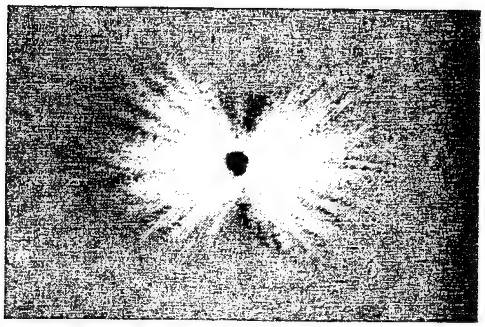
Figure 3. Scattering Patterns from the Photomicrographs in Figure 2.

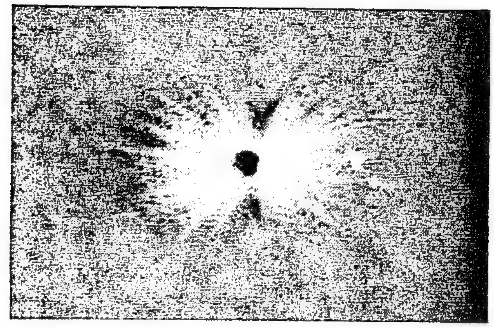


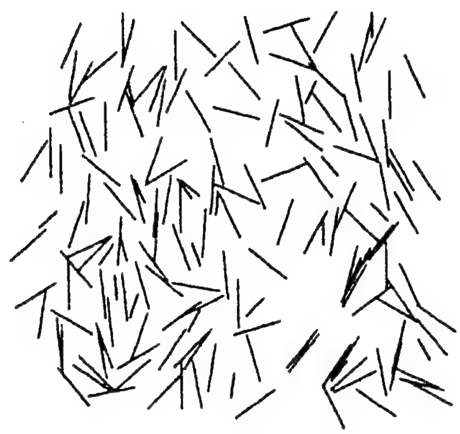
a) orientation, f=.000

b) orientation, f=.142

Figure 4. Computer Drawn Fiber Distribution and the Obtained Scattering Patterns



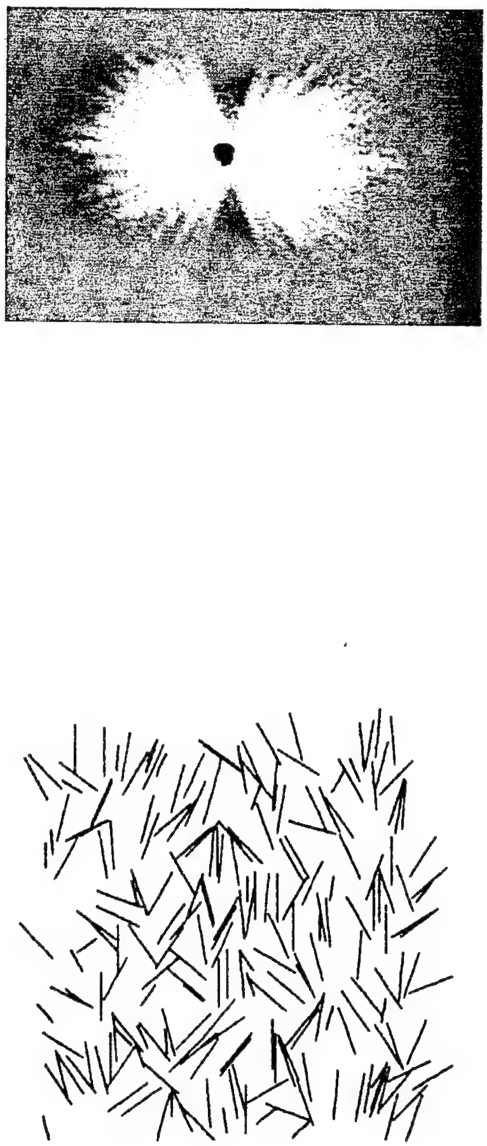




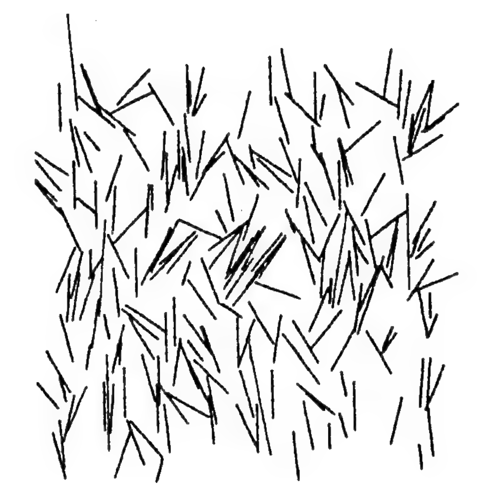
c) orientation, f=.233

d) orientation, f=.329

Figure 4. (cont.)

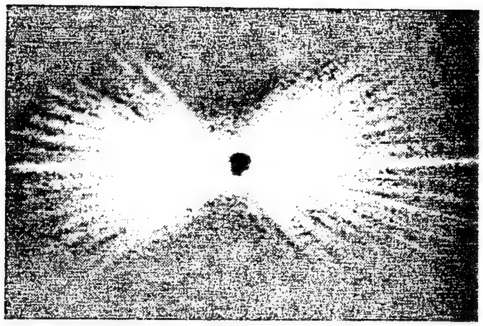


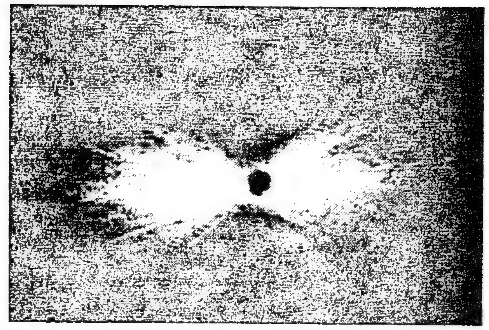
e) orientation, f=.428

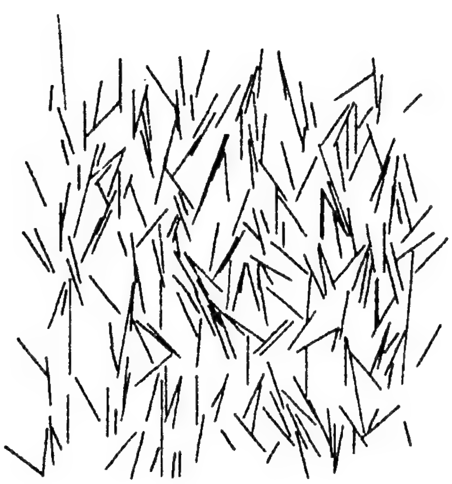


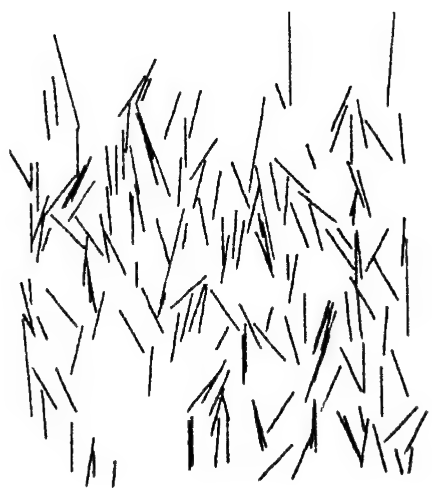
f) orientation, f=.526

Figure 4. (cont.)





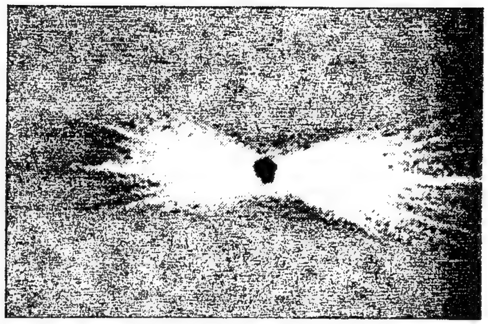


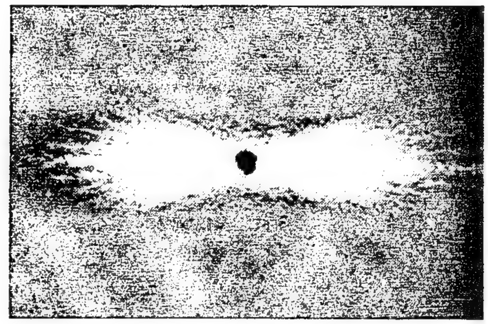


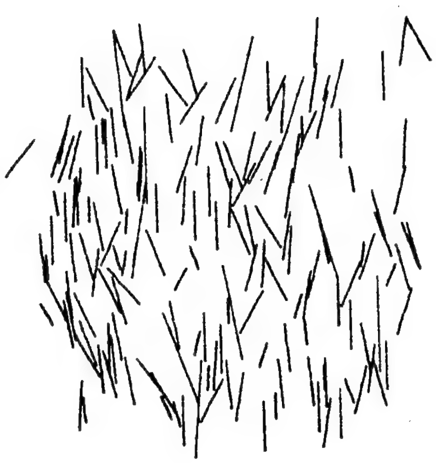
g) orientation, f=.625

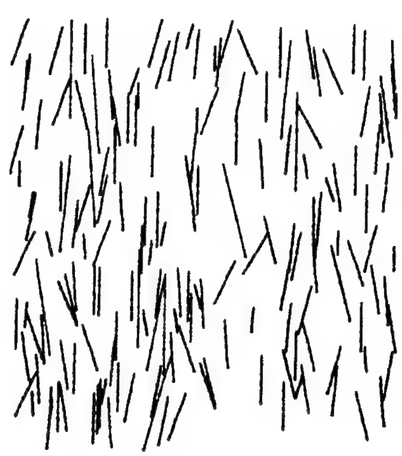
h) orientation, f=.722

Figure 4. (cont.)









i) orientation, f=.818

j) orientation, f=.913

Figure 4. (cont.)

fiber orientation, it was decided to use a computer model to "draw" various states of fiber orientation. The assumptions which were used in developing f_p, viz. the fibers are contained in the plane of the material, were ideally suited to this type of model.

Using the definition of f_p, a computer program was written which would accept as input the desired state of orientation. A random number generation scheme was used to locate the endpoints of each fiber. Before an individual fiber was drawn, however, a check was made to insure that it would not intersect with any "fiber" line which had been drawn previously. This stipulation was introduced to avoid two fibers occupying the same space in the plane.

One of the benefits of using the computer model was that various distributions of fibers which all had the same value of f could be investigated. This assured that the scattering patterns were indeed dependent on the value of f_p and not the entire distribution function. The distribution function which was used was a one parameter model suggested by McCullough[5] based on the data of Tock and McMackins[6]. The form of the distribution was

$$n(\phi) = a*\cos^k\phi$$

where k is the parameter which controls the orientation and a is found from the constraint

$$\int_0^{\pi/2} n(\phi)\, d\phi = 1$$

To investigate the entire range of values of the orientation parameter, computer drawn fiber distributions were constructed at intervals of f_p of approximately .1. These distributions and the resulting scattering patterns are presented in Figure 4.

In Figure 4a, a state of random orientation, the scattering pattern is spherical with no apparent separation of the lobes. As the orientation is increased (Figures 4b through 4j) the separation between the lobes increases with increasing orientation. A slight discrepancy in this trend is found in Figure 4c. Here it should be noted that the total number of fibers in the computer drawn distribution is less than in any of the other Figures. Since the total intensity of the scattering pattern is a function of the total number of fibers, this pattern would have been less intense than the others and hence the region between the lobes would not appear in the photograph of Figure 4c. This result points out the importance of obtaining a consistent scattering intensity.

To further illustrate the affect of fiber orientation on the separation of the lobes, the angle between the lobes was measured. These results are presented in Table 1 and graphically in Figure 5. In Table 1 and Figure 5, the data from Figure 4c has been omitted for the reason previously

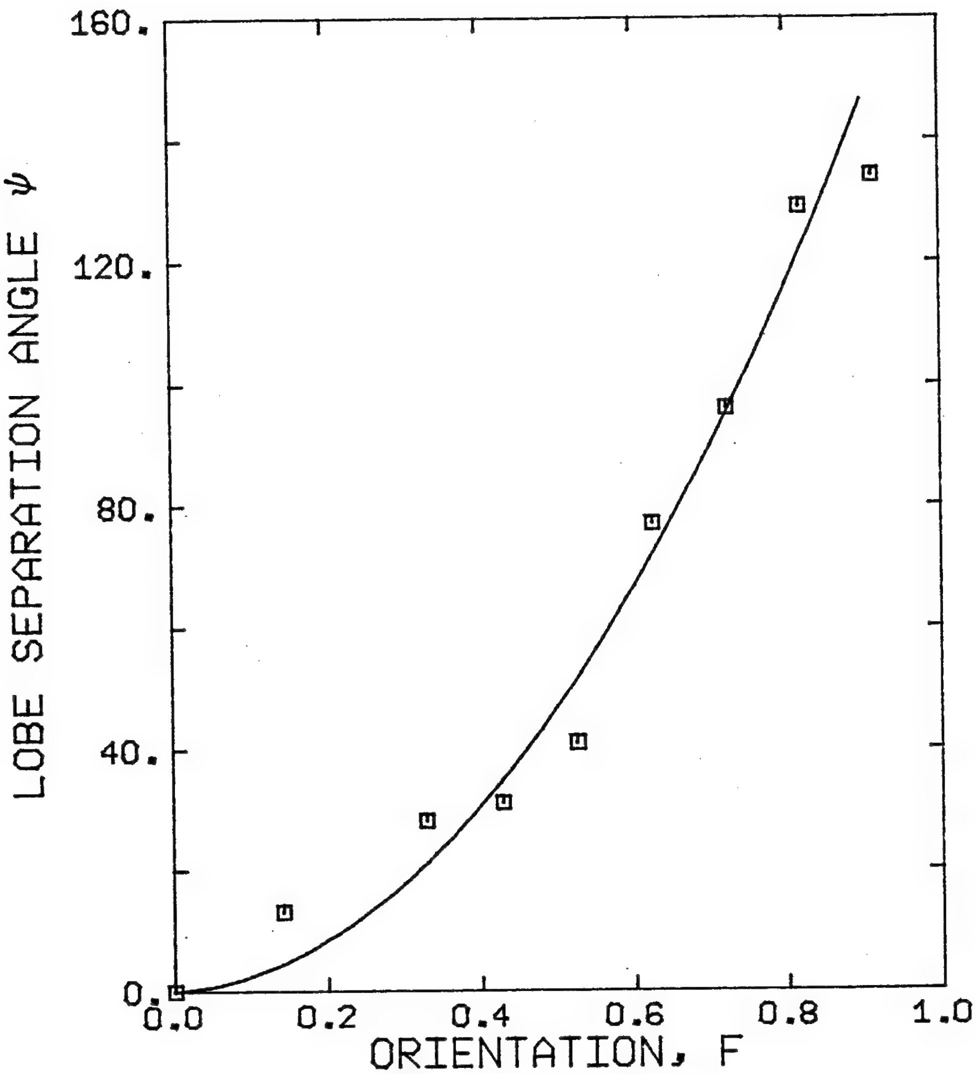


Figure 5. Angular Lobe Separation Data from Computer Drawn Fiber Distributions

Orientation, f	Separation Angle ψ (degrees)
.000	0
.142	13
.329	28
.428	31
.526	41
.625	77
.722	96
.818	129
.913	134

Table 1. Angular Lobe Separation Data from Computer Drawn Fiber Distributions

discussed. The curve drawn in Figure 5 is the least squares fit of the data with the constraints:

1) it pass through the origin
2) it pass through 180 degrees at f_p equal one
3) it be of order two or less

The equation for the curve is

$$\psi = f_p(172*f_p + 8)$$

where ψ is the angular separation, in degrees, between the lobes. The details of the regression analysis are contained in Appendix IV.

Figure 5 provides a calibration curve for obtaining the orientation parameter, f_p, from observed scattering data. For example, the fiber orientation of Figure 2 responsible for the scattering patterns shown in Figure 3 corresponds to f_p values of approximately .9 and .6 respectively. The accuracy of this technique can be improved by the use of

normalized optical density tracings so that a consistent scattering level is used as a reference for the lobe angle. Further improvements can be obtained through the use of photomultipliers.

Conclusions

This study demonstrates that light scattering techniques can be used to obtain orientation parameters in a short fiber composite. The remaining drawbacks are the need to section and polish the sample, but these are not exceptionally time consuming processes. The benefits of this method are the relative ease with which it can be applied and the ease of interpretation of the results.

Future Work

This preliminary investigation by no means completes the study of measuring fiber orientation. Several objectives for future studies include:

1) develop a theoretical analysis for the scattering patterns obtained from short fiber composites

2) use small angle light scattering to investigate the "bundling" of fibers in sheet molding compounds

3) investigate the features of the scattering patterns associated with the distribution of fiber lengths

4) develop a nondestructive test method for evaluating fiber orientation

The first three goals listed above are direct extensions of the current work. It is expected that the results from developed light scattering theory can be adapted to the technique discussed here. The use of small angle light scattering will require minor modifications of the technique.

However, it is the last goal listed that presents the major challenge. Hopefully, by using the results of the light scattering measurements to develop a "calibration curve", an alternate method of measuring fiber orientation can be developed. One of the possible alternate methods is measuring the sonic modulus of the composite. The sonic modulus should be dependent on fiber orientation and if a calibration curve could be constructed, this would provide a quick, nondestructive method for measuring f_p and f_a.

Acknowledgements

The help and instruction of Dr. Mark Sharnoff, Professor, Department of Physics, University of Delaware in the use of his light scattering equipment and for his consultation is greatly appreciated.

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Appendix I

Photographic Technique To Produce Diffraction Mask

The production of the diffraction mask required two separate photographic steps. First, a photomicrograph of the composite material whose fiber orientation was to be determined had to be made. Second, the photomicrograph had to be reduced in size and a high constrast negative, which would serve as the diffraction grating, had to be produced. These two steps are described in detail in this Appendix.

Preparation of the Photomicrograph

The first step in preparing the photomicrograph was the sectioning and polishing of the composite material. The sectioning was performed using a diamond saw. This allowed the fiber orientation in any region of the sample to be investigated. The sectioned sample was then mounted in an epoxy resin holder to facilitate subsequent polishing. The mounted sample was polished using several different grits of wet sand paper followed by several different polishing powders. The final grade of polishing powder used was one micron. The process of sectioning and polishing is described in a step by step procedure in "Preparation of Sections of Resin Matrix Composites for High Resolution Optical Microscopy" by M. G. Bader, Center for Composite Materials, University of Delaware.

Once the sample had been polished, the photomicrograph could be taken. Polaroid type 55 positive/negative film was used in a reflecting photomicroscope. Magnification was set at 50X. The positive image was purposely overexposed to produce a dark negative which highlighted the difference between the fibers and the surrounding resin. Optimum exposure time was found to be 25 seconds when no filters were used in the microscope. These negatives were rinsed in sodium nitrite for a few minutes to remove the outer film.

Preparation of the Diffraction Mask

To produce the diffraction mask from the photomicrograph negative, a negative image was taken which both reduced the size of the image and also resulted in the fibers being the light portion of the photograph. The photomicrograph negative was placed on a light box with a 35mm camera mounted above. The camera was equipped with a wide angle lens which served to reduce the image. To obtain the correct size diffraction mask, approximately .25 inches square, the camera was located 24 inches above the photomicrograph negative.

The film used for the diffraction mask was Kodalith graphic line film. This type of film was used since it was necessary to obtain a high contrast image using a small grain size film. To insure that an optimum diffraction mask was produced, three exposures of each negative were made.

The "f" stop settting was set at the correct value according to the light exposure reading and one exposure taken at this setting. Additional exposures were made at "f" stop settings one greater than and one less than the correct value to obtain the desired masks.

To develop the Kodalith film, it was first placed in a 35mm tank. The remainder of the process for developing the film at room temperature is detailed below.

Process	Time
Kodalith developer, parts A and B	2.75 min
Stop bath	10 sec
Rapid fixer or	1-2 min
Fixer	2-4 min
Water rinse	10 min

The film was then hung to dry. Individual slides were cut, mounted into slide mounts and the region surrounding the composite was masked using a black, opaque tape. This completed the preparation of the diffraction masks.

Appendix II

Observing the Scattering Pattern

The diffraction masks produced according to the details given in Appendix I, were placed in the laser diffraction apparatus. The beam from the laser was shown through a pinhole to reduce the beam to the size of the diffraction mask. This reduced beam was shown through the diffraction mask and the scattering pattern observed on a screen placed eight feet from the mask. Since the scattering pattern was faint, it was necessary to darken the room to observe the pattern.

Due to the relatively bright intensity of the central, non-diffracted portion of the laser beam, it was desired to block out this portion of the beam before photographing the scattering pattern. This was done by placing a hole in the screen which was the size of the central beam. Behind this hole, a mirror was mounted at a 45 degree angle to the incident beam. This mirror reflected the central beam away from the observer and effectively removed it from the scattering pattern. A schematic diagram of the mirror and screen is shown in Figure II-1.

A 35mm cammera was set on a tripod approximately two feet from the diffraction screen and as close to the central beam as possible without blocking the scattering pattern. For the 100 milliwatt laser used here, an exposure time of

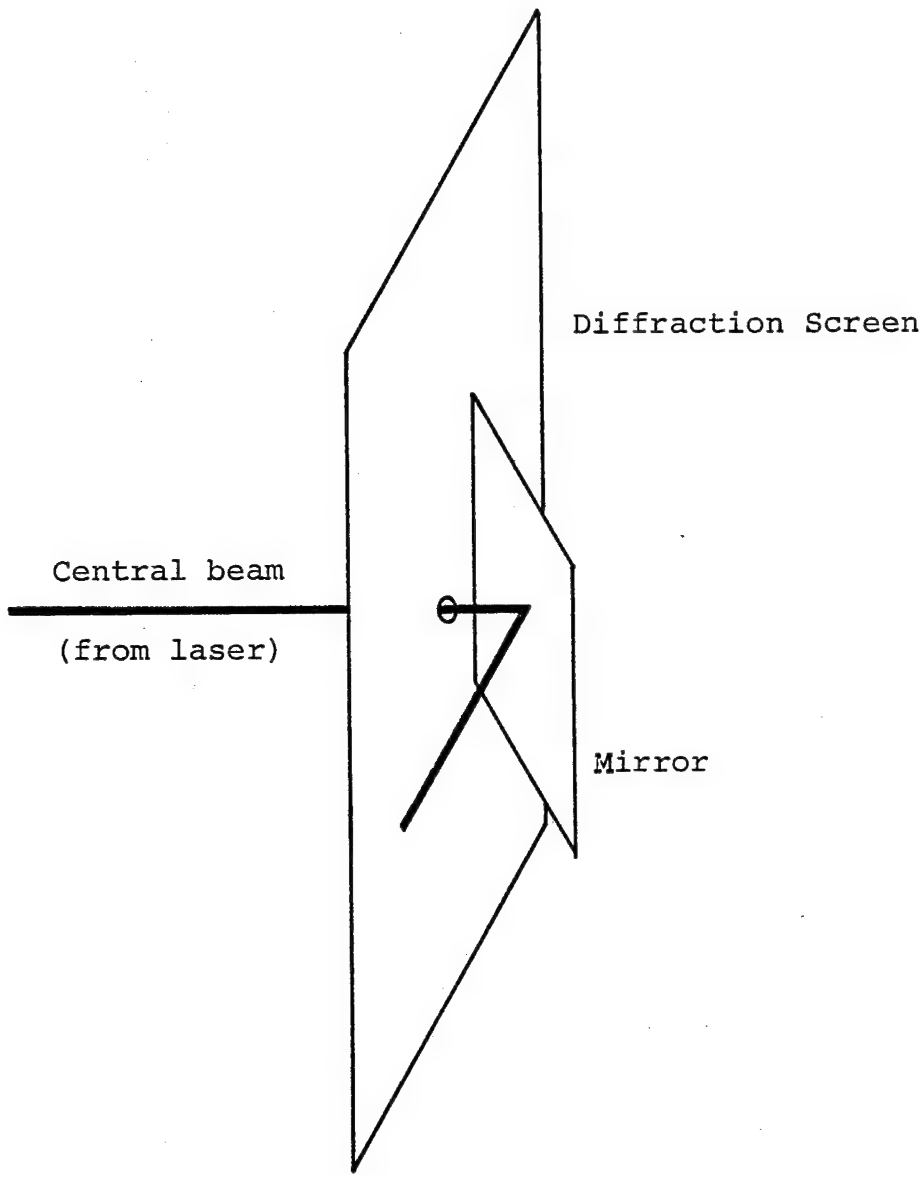

Figure II-1. Schematic Diagram of Reverse of Diffraction Screen

two seconds at an "f" stop setting of 2.8 using ASA 400 black and white print film was found to produce the best results. However, a series of exposures was taken which bracketed this value to insure that an acceptable print was obtained since the intensity of each diffraction pattern varied from mask to mask.

Appendix III

Producing Prints of the Original Sample and the Diffraction Pattern

To produce prints of the original photomicrograph, a standard black and white photographic paper, such as Kodak polycontrast double weight paper, was used. An image the same size as the original, 3.5 by 4.5 inches, was obtained by placing the Polaroid negative directly on top of the photographic paper. This was then exposed to light for approximately 15 seconds (this time is approximate and needs to be checked to obtain the optimum exposure). The paper was then processed in the following manner.

Process	Time
Developer	1.75 min
Stop bath	30 sec
Fix bath	3 min
Water wash	1 min
Fix clearing bath	3 min
Water wash	20 min

Images of the scattering pattern were obtained by first placing the negative obtained from the instructions in Appendix II in an enlarger. The size of the image was adjusted by varying the distance between the enlarger and the paper. A ten second exposure with the aperture of the enlarger set at "f" 8 was usually found to be satisfactory.

This was taken as a starting point and if necessary additional exposures were made if the first attempt did not yield a satisfactory print. The paper was processed as described above. An excellent description of the process of developing photographic prints is given in "Photography" by John and Barbra Upton, Educational Associates, 1975.

Appendix IV

Regression Analysis

When using linear regression, the intent is to find the "best fit" parameters to an equation of the form,

$$y_p = b_0 + b_1 x + b_2 x^2 + \ . \ . \ . \ + b_n x^n$$

where the b_i, $i=0,1,...,n$, are the parameters to be determined, x is the independent variable and y_p is the dependent variable. In this context, the angle between the lobes of the scattering pattern was the dependent variable and the fiber orientation, as measured by the Herman's orientation parameter f, was the independent variable. For the current problem, it was decided to truncate the regression equation after the third term.

Two constraints were placed on the coefficients which were determined from the regression model. First, the function y_p had to pass through the origin since randomly oriented fibers produced a uniform, circular scattering pattern. This required that the coefficient b_0 be identically zero. Secondly, when the fibers were perfectly aligned, the estimated separation between the lobes was to be 180 degrees. This constraint was expressed as

$$b_1 + b_2 - 180 = 0$$

The definition of "best fit" was the minimum of the function

$$F = \Sigma(y - y_p)^2$$

This is the usual least squares regression criterion. To minimize F, the derivative of F with respect to each of the coefficients was taken and set equal to zero. This resulted in the following equations.

$$\partial F/\partial b_1 = -2\,\Sigma yx + 2b_1\Sigma x^2 + 2b_2\Sigma x^3$$

$$\partial F/\partial b_2 = -2\,\Sigma yx^2 + 2b_1\Sigma x^3 + 2b_2\Sigma x^4$$

The remaining constraint was introduced into the problem at this point using a Lagrangian multiplier. The equations which resulted are given below.

$$-2\,\Sigma yx + 2b_1\Sigma x^2 + 2b_2\Sigma x^3 + \lambda(b_1 + b_2 - 180) = 0$$

$$-2\,\Sigma yx^2 + 2b_1\Sigma x^3 + 2b_2\Sigma x^4 + \lambda(b_1 + b_2 - 180) = 0$$

$$b_1 + b_2 - 180 = 0$$

In these expressions λ is the Lagrangian multiplier. Eliminating λ from the above expressions and then solving for b_2 yields

$$b_2 = \frac{\Sigma yx^2 - \Sigma yx + 180(\Sigma x^2 - \Sigma x^3)}{\Sigma x^4 - 2\,\Sigma x^3 + \Sigma x^2}$$

The remaining coefficient, b_1, was found from

$$b_1 = 180 - b_2$$

To estimate the adequacy of the model just developed, the standard correlation coefficient was calculated. Since the two constraints had been placed on the model, it was necessary to use the most general definition of the correlation coefficient, viz.

$$r^2 = 1 - SSE/Syy$$

where SSE and Syy are given by

$$SSE = \Sigma(y - y_p)^2$$

$$Syy = \Sigma(y - \bar{y})^2$$

and $\bar{y}$ is the average of the measured y values. The calculated correlation coefficient was .97. This indicated that the model accounted for 97% of the variation in the observed data. For this reason, it was determined that the model described here adequately fit the experimental data and that the induced constraints were reasonable.